

Quantitative Mass Transfer in Coastal Sediments During Early Diagenesis: Effects of Biological Transport, Mineralogy, and Fabric

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Award Number: N00014-01-1-0599
<http://www.geology.wmich.edu/koretsky/homepage/homepage.html>
http://www.geo.uu.nl/Research/Geochemistry/P_vCappellen.html

LONG-TERM GOALS

The long-term goal of this project is to develop multicomponent reactive transport models that can be used to accurately describe solute transport in temporally and spatially heterogeneous, organic-rich sediments. To achieve this goal, we have developed a model that can be used to obtain unbiased 1D quantitative descriptions of biologically-enhanced solute transport (bioirrigation due to macrofaunal activities) from chemical data. We have also developed a quantitative 1D description of bioirrigation that relies on ecological data (i.e., information regarding burrow networks) and requires minimal chemical data. A long-term goal of the project is to use these models to assess the spatial and temporal variability of biologically-enhanced solute transport in a wide range of sedimentary environments and to incorporate these models into existing 1D coupled reactive-transport models. The results of this work will also be used to develop flexible 3D models of enhanced solute transport. These flexible 3D models will be useful for investigation of many spatially and temporally invariant processes, including not only bioirrigation, but also, for example, the effects of macrophytes on solute transport in near-shore sediments.

OBJECTIVES

The primary fiscal year objective was to complete development of the 1D stochastic bioirrigation model and to use the model to assess bioirrigation intensity in a series of saltmarsh sites located at Sapelo Island, GA and to disseminate and publish the results of this work. A secondary objective was to initiate the development of 3D models of bioirrigation.

APPROACH

A stochastic model of burrow distributions was developed to function as a link between ecological data and nonlocal bioirrigation coefficients. This approach allows the extreme spatial and temporal

Report Documentation Page				Form Approved OMB No. 0704-0188	
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1. REPORT DATE 30 SEP 2003		2. REPORT TYPE		3. DATES COVERED 00-00-2003 to 00-00-2003	
4. TITLE AND SUBTITLE Quantitative Mass Transfer in Coastal Sediments During Early Diagenesis: Effects of Biological Transport, Mineralogy, and Fabric				5a. CONTRACT NUMBER	
				5b. GRANT NUMBER	
				5c. PROGRAM ELEMENT NUMBER	
6. AUTHOR(S)				5d. PROJECT NUMBER	
				5e. TASK NUMBER	
				5f. WORK UNIT NUMBER	
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Western Michigan University,,Department of Geosciences,,Kalamazoo,,MI,49008				8. PERFORMING ORGANIZATION REPORT NUMBER	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)				10. SPONSOR/MONITOR'S ACRONYM(S)	
				11. SPONSOR/MONITOR'S REPORT NUMBER(S)	
12. DISTRIBUTION/AVAILABILITY STATEMENT Approved for public release; distribution unlimited					
13. SUPPLEMENTARY NOTES					
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15. SUBJECT TERMS					
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT Same as Report (SAR)	18. NUMBER OF PAGES 8	19a. NAME OF RESPONSIBLE PERSON
a. REPORT unclassified	b. ABSTRACT unclassified	c. THIS PAGE unclassified			

heterogeneity of nearshore deposition environments to be considered explicitly in constraining solute transport via bioirrigation. The basic approach is to develop probabilistic descriptions of burrow morphologies and densities and to use these to simulate 3D macrofaunal burrow networks. Probabilistic densities are derived for individual organisms based on resin cast and X-radiography data (e.g., Furukawa et al., 2001). The volume occupied by the burrows is used to calculate the sediment porosity due to burrow networks as a function of depth, and the burrow surface area as a function of depth is used to derive nonlocal bioirrigation coefficient profiles according to:

$$\alpha_i = \frac{SA_{\text{slice}}}{V_{\text{slice}}} \frac{D_i}{\bar{r} - r_1}$$

where α_i is the nonlocal exchange coefficient, SA_{slice} is the total burrow surface area over a discrete depth interval, V_{slice} is the corresponding volume of sediment, D_i is the effective molecular diffusion coefficient of solute i , r_1 is the radii of burrows assuming that they are cylindrical, and $\bar{r} - r_1$ is the reactive length scale (L), that is, the distance from the burrow at which solute i reaches the bulk sediment concentration ($C_{\text{average}}(x)$) (Aller, 1980; Boudreau, 1984). \bar{r} is constrained using (1) direct measurements across burrow walls ($\bar{r} = r_1 + L$) (b) measurements across the sediment-water interface (SWI) corrected for the radial geometry of burrows ($\bar{r} = \sqrt{r_1^2(x) + 2r_1(x)L}$) or (c) by calculation using a depth-dependent measured or estimated reaction rate, $R(x)$, for solute i , according to

$$L = \sqrt{\frac{D_i \cdot (C_o - C_{\text{average}}(x))}{R(x)}}$$

where C_o is the concentration of solute i at the SWI.

The stochastic model was used to derive bioirrigation coefficients from the sediment water to a depth of up to 50cm at creek bank and vegetated ponded marsh sites in a saltmarsh at Sapelo Island, GA (Koretsky et al., 2002; Koretsky et al., 2003a). Crab, polychaete worm and shrimp burrow densities and network sizes and shapes were estimated using literature data (e.g., Teal, 1958; Allen and Curran, 1974; Basan and Frey, 1977). Coefficients were derived either by assuming zero order kinetics for sulfate reduction or by estimating a reactive length scale (L) from reported O_2 fluxes (Cai et al., 1999).

Development of a 3D finite element has been initiated by principal investigator Van Cappellen in close collaboration with Dr. Christof Meile, in order to simulate reaction and transport in spatially and temporally heterogeneous sediments (Meile et al., 2003). The fluid velocity (q) is related to the pressure field by:

$$q = \phi v - \frac{k}{\mu} (\nabla p - \rho g)$$

where ρ , ϕ , μ , k , p and g are density, porosity, viscosity, permeability, pressure and gravitational acceleration the distribution of dissolved chemical species is governed by:

$$\frac{\partial \phi C_m}{\partial t} = \nabla \cdot (D^* \nabla C_m) - \nabla \cdot (\phi v C_m) + \phi R_m$$

where C_m , t , D^* , R_m are the mass of species m per unit volume pore water, time, the diffusion tensor and the sum of all reaction rates relevant to C_m . An explicit time stepping scheme is used, where advection is calculated first, followed by concentration fields (Meile et al., 2003).

WORK COMPLETED

The stochastic biorrigation model has been developed and published (Koretsky et al., 2002). The model has been thoroughly tested using mesocosm data. Bioirrigation coefficients derived using the stochastic model have been compared to coefficients estimated independently using the chemically-based inverse model. Results from the two models are in close agreement (Koretsky et al., 2002). The model has been applied to three field sites forming a transect through a saltmarsh on Sapelo Island, GA. The results have been used to interpret spatial and temporal variations in pore water and solid phase chemistry at these sites (Koretsky et al., 2003a). The 3D model has been used to investigate the distribution of oxygen and nitrate in a model system with a single, continuously-flushed U-shaped burrow (Meile et al., 2003).

RESULTS

Bioirrigation coefficients derived using ecological data with the stochastic model were shown to be in good agreement with independently derived coefficients derived using the chemically-based inverse model (Fig. 1). This is important, because it suggests that the stochastic model can be used to reliably predict bioirrigation coefficients even for environments for which minimal chemical data is available.

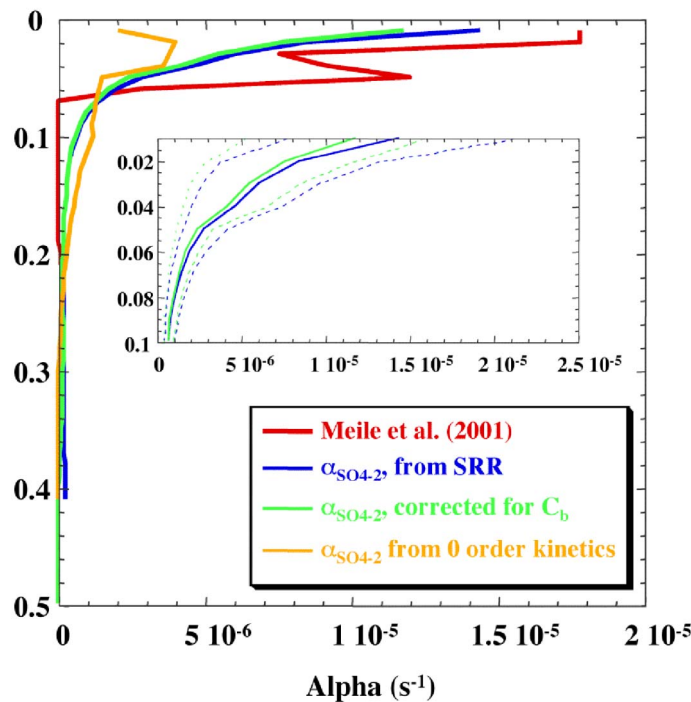


Figure 1. Bioirrigation coefficients as a function of depth (s-1) derived using inverse modeling of chemical data (red) and derived using the stochastic model with either measured sulfate reduction rates corrected for imperfect burrow flushing (green), using measured sulfate reduction rates uncorrected for imperfect flushing (blue) or assuming zero order kinetics for sulfate reduction (orange). [Bioirrigation coefficients from all models decrease from the sediment water interface to become 0 at between 9 and 20 cm depth. The stochastic model results based on measured sulfate reduction rates are in especially good agreement with those estimated using inverse modeling of chemical data.]

Bioirrigation coefficients were derived for three saltmarsh sites forming a transect from a large tidal creek into a vegetated ponded marsh. It was demonstrated quantitatively that bioirrigation is more intense and extends deeper into the sediments at the unvegetated creek bank site, compared to the vegetated ponded marsh (Fig. 2). These results were used to explain the more compressed redox stratification observed in the ponded marsh sediments (Koretsky et al., 2003a).

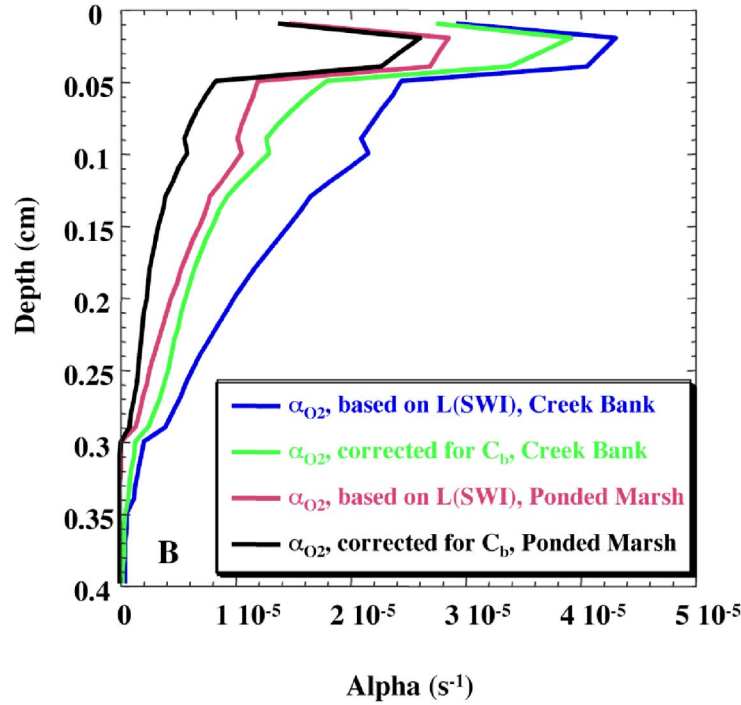


Figure 2. Bioirrigation coefficients as a function of depth (s^{-1}) derived using dissolved oxygen at the creek bank (green) or vegetated bonded marsh (black) with correction for solute depletion within burrows or without correction for solute depletion at the creek bank (blue) and ponded marsh (purple). [Bioirrigation coefficients increase from the sediment water interface to approximately 2.5 cm depth at both sites, then decrease to zero by 30 cm depth at the ponded marsh or 40 cm depth at the creek bank. Bioirrigation coefficients are larger at the creek bank at all depths and are larger without correction for solute depletion.]

The 3D reactive transport model developed primarily by Christof Meile (Meile et al., 2003) quantitatively demonstrates that the presence of flushed burrows in sediments can create significantly increase the penetration of oxygen or nitrate into the sediment column (Fig. 3).

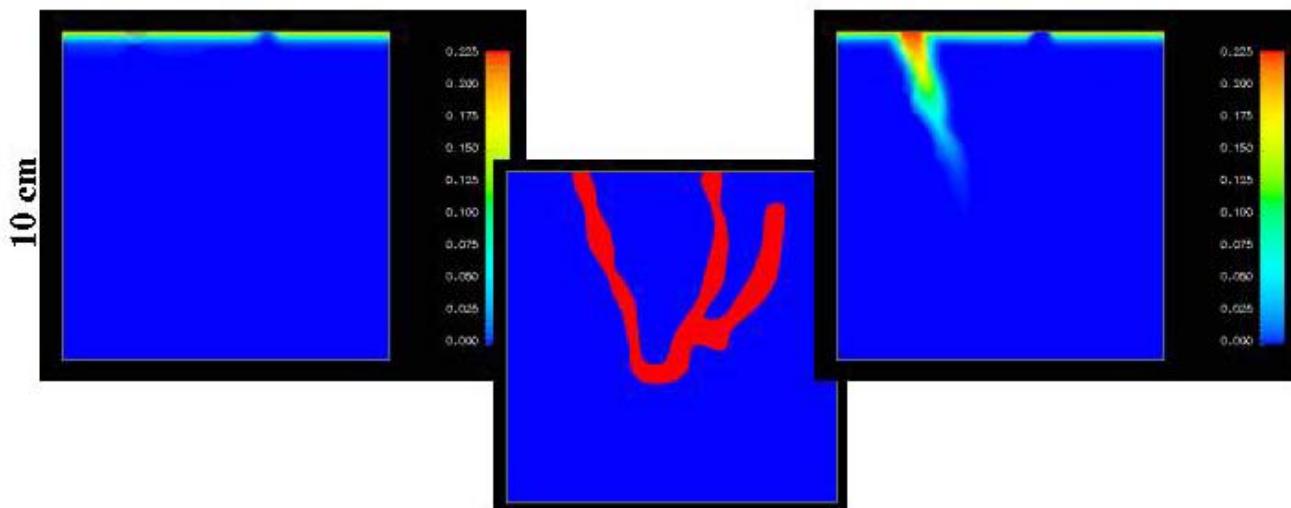


Figure 3. 3D model simulations illustrating the influence of macrofaunal burrow flushing on enhanced dissolved oxygen transport as a function of depth. Middle panel illustrates U-shaped burrow extending to depth of 5 cm, with side branch, used for the simulation. Panel to the left shows oxygen concentrations (higher concentrations shown in red, lower concentrations shown as blue) in a homogenous, burrow-free sediment. Panel to the right shows oxygen concentration field in the presence of continuously flushed burrow. [Oxygen penetration depth is limited to upper mm in unburrowed sediments, but oxygen concentrations are measureable at depths of 2.5 cm or more in the case of flushed U-shaped burrow.]

IMPACT/APPLICATIONS

The stochastic and inverse bioirrigation models developed in this study will be useful for assessing enhanced solute transport, especially in spatially and temporally heterogeneous, organic-rich coastal sediments. Bioirrigation coefficients derived from these models can be directly incorporated into existing reactive transport models, such as STEADYSED (Van Cappellen and Wang, 1996) or into new more flexible reactive transport models that are currently being developed (e.g., Regnier et al., 2002; Meysman et al., 2003a; b). The 3D model initiated here will shed light on temporal and spatial variability of enhanced solute transport due to macrofaunal and macrophyte activity in coastal sediments.

RELATED PROJECTS

Seasonal pore water profiles of dissolved species (e.g., Fe(II)/Fe(III), SO_4^{2-} , H_2S , PO_4^{3-} , NH_3 and Mn) have been measured in saltmarshes at Sapelo Island, GA and at the Scheldt Estuary (Netherlands and Belgium). The seasonal oscillation of microbial community structure at these same sites is being studied in collaboration with T. DiChristina and C. Moore (Georgia Institute of Technology). Interpretation of spatial and temporal oscillations in the saltmarsh geochemistry and microbiology has been greatly aided by results of inverse and stochastic models developed in this study (Koretsky et al.,

2003b). The relative compression of redox zones in these sediments has been shown to strongly depend on the depth and intensity of bioirrigation (Koretsky et al., 2003a).

The global influence of enhanced solute transport on oxygen and phosphate fluxes has been estimated (Meile and Van Cappellen, 2003). Nonlocal mass transfer bioirrigation coefficients were obtained through comparisons of measured total oxygen fluxes from benthic chambers and benthic fluxes due to molecular diffusion, estimated from high resolution pore water profiles. The results of this study indicate that continental shelves, in particular, are strongly influenced by enhanced solute transport processes.

The stochastic, ecologically-based model developed in this study may prove to be useful for studies of solute transport in paleosediments. Well-preserved trace fossils may be useful for constraining burrow network parameters, and trace metals surrounding such fossils (e.g. Harding and Risk, 1986; Over, 1990) may be useful as proxies that can be used to constrain the reactive length scale (L). These hypotheses are being pursued in collaboration with S. Walker (paleontologist, University of Georgia).

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